

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
24 April 2003 (24.04.2003)

PCT

(10) International Publication Number  
**WO 03/033612 A1**

- (51) International Patent Classification<sup>7</sup>: C09J 123/10      (81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, OM, PH, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZM, ZW.
- (21) International Application Number: PCT/US02/26297
- (22) International Filing Date: 16 August 2002 (16.08.2002)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data:  
09/982,641      18 October 2001 (18.10.2001) US
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- (84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

— with international search report

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

A1

WO 03/033612

(54) Title: HOT MELT ADHESIVE COMPOSITION BASED ON A BLEND OF AMORPHOUS POLY-\$G(A)-OLEFIN AND SYNDIOTACTIC POLYPROPYLENE

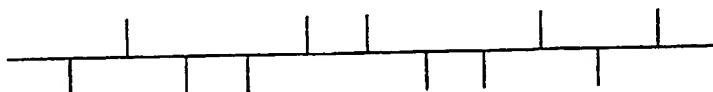
(57) Abstract: A hot melt adhesive composition based on a polymer blend of syndiotactic polypropylene (SPP) and atactic poly- $\alpha$ -olefin (APO). The composition contains about 15%-80% by weight of the SPP/APO blend, about 15%-65% by weight of a compatible tackifier, about 0%-35% by weight of a plasticizer, about 0%-3% by weight of a stabilizer, and optionally, about 0%-30% by weight of a wax. The adhesive composition may be used in a number of applications such as, for example, in disposable non-woven hygienic articles, paper converting, flexible packaging, wood working, carton and case sealing, labeling and other assembly applications.

case, the substrate is brought into intimate contact with a hot die under pressure. The temperature of the die must be maintained well above the melting point of the adhesive, which is typically between 150 and 200 °C. For some applications, particularly for manufacturing nonwoven articles, bonding of delicate and heat sensitive substrates, such as thin gauge polyethylene films, is often involved. Direct contact between the film and the die, in these cases, must be avoided to prevent the film from burning or distorting. Several application methods have been developed through which a hot melt adhesive can be spray coated with the aid of compressed air onto a substrate from a distance. These non-contact coating techniques include spiral spray and various forms of melt-blown methods. Direct contact between the coating head and the substrate is thus eliminated. All the coating techniques herein described above are well known to those skilled in the art and commercial equipment is readily available.

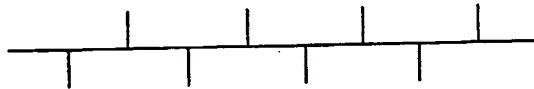
The spray coating techniques, however, pose stringent requirements on hot melt adhesives. The viscosity of the adhesives must be sufficiently low, usually in the range of 2,000 to 30,000 cP, preferably in the range of 2,000 to 15,000 cP, at the application temperature. Many other physical factors, especially the rheological properties of the adhesive, come into play in determining the sprayability of a hot melt. The majority of commercial hot melt products do not lend themselves to spray applications. There are no accepted theoretical models or guidelines to predict sprayability and it must be determined empirically with application equipment.

In accordance with the present invention, it has been found that a mixture comprising a polymer blend of SPP and APAO, a tackifying resin, a plasticizer and, optionally, a synthetic polyolefin wax or petroleum wax provides a sprayable hot melt adhesive composition. The composition has novel combinations of properties including toughness, low or no shrinkage, high cohesive strength, low viscosity, excellent heat stability and good adhesion to a variety of substrates. The composition of the present invention is particularly useful in assembly of

hexene. In contrast to the regular structures in IPP or SPP, APAOs have atactic molecular chains with the methyl groups on the successive monomeric units sterically randomly distributed on both sides of the hypothetical plane through the polymer chain. The stereo configuration of the atactic APAO molecular chain can 5 be illustrated graphically by using the following Fisher projection formula:



The stereo chain structure of SPP is uniquely different from that of IPP and from that of APAO. In contrast to the isotactic chain configuration of IPP and the 10 atactic chain configuration of APAO, the stereochemistry of SPP can be described as having the tertiary methyl groups of successive monomer units along the chain alternatively disposed on each side of the hypothetical plane. The stereo configuration of SPP can be depicted below:



15 The stereo configuration of polypropylene can also be characterized quantitatively through C-13 NMR. In NMR nomenclature, a "meso" dyad of successive methyl groups on the same side of the plane, as in the case of IPP, is represented by the letter m. A "racemic" dyad of successive methyl groups on the 20 opposite sides of the plane, as in the case of SPP, is represented by the letter r. The percentage of m or r defines the degree of polymer tacticity with the sum of m and r equal to 100%. Thus, a perfect isotactic polypropylene will have 100% m dyad, whereas a perfect syndiotactic polypropylene will have 100% r dyad. This unique 25 stereochemical structure of SPP results in an unusual and desirable combination of physical and mechanical properties such as low density, low melting point, flexibility and toughness.

The syndiotactic polymers used in the present invention preferably have an r value equal to or greater than 70%. The polymers having an r value greater than

PB and the conventional IPP, very often such modifications not only failed to rectify the problems, but also led to adverse side effects.

For example, Trotter et al, in U.S. Patent No. 4,022,728, describes a hot melt pressure sensitive composition comprising a mixture of APAOs, a low molecular weight substantially amorphous elastomer, a liquid tackifier and a conventional crystalline polypropylene (IPP) in the amount of up to 2% by weight. It is claimed that the composition provides good adhesive properties at low temperatures.

Meyer et al, in U.S. Patent 4,120,916, discloses hot melt adhesive compositions comprising a blend of low molecular weight polyethylene, low molecular weight conventional propylene containing polymer and APAO. These adhesive compositions are said to offer short open time and to be useful for bonding of paraffin modified corrugated board.

Lakshmanan et al, in U.S. Patent No. 4,761,450, discloses a polymer blend useful as hot melt adhesive comprising a low density ethylene polymer, a copolymer of butene-1 with ethylene or propylene, a hydrocarbon tackifier and a low molecular weight polymer consisting of a low molecular weight liquid polybutene, an amorphous polypropylene and mixtures thereof.

Lakshmanan et al, in U.S. Patent No. 5,478,891, also discloses blend compositions containing (a) a high molecular weight copolymer of ethylene with an  $\alpha$ -olefin having at least 4 carbons and (b) an amorphous polypropylene or amorphous polyolefin. The components of the blends are described as having molecular weight range between 300 to 6000. The polymer blends are claimed to be useful for hot melt adhesives, coatings, sealants, asphalt modifiers and plastic additives.

Ryan discloses in U.S. Patent No. 5,747,573 an APAO based hot melt adhesive composition useful for bonding plastics and metallized foil containers. The adhesive composition contains a blend of APAO, a solid benzoate plasticizer and a hydrocarbon tackifier.

In a prior U.S. Patent 5,317,070, Brant et al disclosed a hot melt adhesive based on tackified SPP having a polymer chain of at least 80% racemic dyads and having a melting point of about 100 to 180 °C. The adhesive is claimed to have good open times between the application of the adhesive and the formation of the joint. This type of tackified SPP usually lacks flexibility and toughness, and therefore, will also have poor bond strength and poor impact resistance.

5 Furthermore, SPP exhibits an inherent shrinkage problem when it transforms from liquid to solid crystalline state. The shrinkage often causes stress concentration at adhesive/substrate interfaces, and consequently, catastrophic bond failure.

10 It therefore would be advantageous to provide a hot melt adhesive that will overcome the shortcomings of the prior art adhesives herein mentioned above. It is found in the present invention that a synergistic effect exists in an SPP/APAO blend hot melt composition. Due to this synergy, an SPP/APAO polymer blend possess a unique combination of properties which the prior art APAO and SPP based hot melt systems have failed to offer.

#### SUMMARY OF THE INVENTION

The present invention is directed to a hot melt adhesive composition based on a polymer blend of the SPP and APAO. The adhesive comprises, in addition to the SPP/APAO blend, a tackifying resin, an optional plasticizer and an optional wax as the primary ingredients. The composition of the present invention takes advantage of complimentary properties between SPP and APAO and has overcome the shortcomings of the prior art APAO blend adhesives and tackified SPPs. The composition of the present invention provides well balanced properties of tensile strength, toughness, flexibility and adhesion. It shows complete compatibility, excellent heat stability, adjustable open time, improved cohesive strength, low viscosity, low shrinkage upon solidification, low or no tack when set, and good processability with conventional coating equipment. In particular, the present invention leads to an adhesive composition that is well suited for a variety of spray

nonwoven fabric, or between two nonwoven fabrics. This type of adhesive can be formulated to have dual functions for both elastic attachment and construction.

Another objective of the present invention is to provide a hot melt adhesive for carton and case sealing to provide a strong bond at sub-ambient temperatures.

5 Due to its toughness and flexibility, an SPP/APAO based hot melt is advantageous for low temperature applications. The adhesive of the present invention would offer fiber tear bond at the ambient temperature.

The hot melt adhesive composition of the present invention comprises as components thereof a mixture of the following ingredients:

- 10 a. A blend of syndiotactic polypropylene (SPP) polymer and atactic poly- $\alpha$ -olefin (APAO) in the amount of about 15% to 80% by weight, preferably in the amount of about 25% to 60% by weight, and most preferably in an amount of from about 35% to 55% by weight, said blend having a SPP to APAO ratio from about 10% by weight of SPP and 90% by weight of APAO to about 90% by weight of SPP and 10% by weight of APAO; said blend having a most preferred ratio of 20% SPP and 80% APAO; said SPP having a density of about 0.85 g/cc to 0.92 g/cc and a melt flow rate of equal to or greater than 1 g/10 min and a  $r$  value equal to or greater than 70%; and said APAO having a density of about 0.85 g/cc to 0.89 g/cc and a glass transition temperature ( $T_g$ ) of from about -5 to -40 °C and a weight average molecular weight ( $M_w$ ) of from about 4,000 g/mol to about 150,000 g/mol.;
- 15 b. A compatible tackifier in the amount of 15% by weight to 65% by weight, preferably in the amount of 30% by weight to 50% by weight;
- 20 c. About 0% to 35% by weight, preferably about 5% to 30% by weight, of a plasticizer;

weight, preferably about 5% to 30% by weight, of plasticizer, about 0% to 30% by weight, preferably about 0% to 18% by weight, of wax and about 0% to 3% by weight of stabilizer or antioxidant. Optional components such as filler, colorant, blowing agent, fluorescing agent and the like can be added to the basic composition to modify its properties, as desired.

The hot melt composition of the present invention includes a SPP polymer. The art of preparing SPP polymers have been disclosed in U.S. Patent 3,305,538 and 3,258,455 to Natta et al, U.S. Patent 4,892,851 to Ewen et al, U.S. Patent 5,270,410 to Job, U.S. Patent 5,340,917 to Eckman et al, U.S. Patent and U.S. Patent 5,476,914 to Ewen et al., the entire disclosures of which are hereby incorporated by reference. While syndiotactic propylene homopolymer can be used in the hot melt composition of the present invention, syndiotactic polypropylene copolymers are preferred. Suitable SPP copolymers can be prepared by copolymerization of propylene with other unsaturated olefin monomers containing 2 to 10 carbon atoms, which include, but are not limited to, ethylene, butene-1, pentene-1, 4-methyl pentene-1, hexene-1 and octene-1. These homopolymers and copolymers can be manufactured with any prior art process herein described above. However, the polymers prepared by using method disclosed in U.S. Patent 5,476,914 with metallocene catalyst system are preferred. Compared with other methods, the SPP polymers prepared with metallocene catalyst will have better stereo-regularity, narrower molecular weight distribution and more uniform comonomer distribution, which lead to better physical and mechanical properties and superior processability. The most preferred type of SPP polymers is metallocene catalyzed copolymer of propylene with ethylene or butene-1 having ethylene or butene-1 comonomer content ranging from about 2% to 20% by weight.

The SPP polymers useful in the present invention preferably have a  $r$  value equal to or greater than 70%, more preferably greater than 80% and most preferably greater than 85%. The said polymers generally have a density in a range from about 0.85 g/cc to about 0.90 g/cc and preferably from 0.86 g/cc to 0.88 g/cc at the room

The tackifying resins or tackifiers which are used in the hot melt adhesives of the present invention are those which extend adhesive properties and improve specific adhesion. As used herein, the term "tackifying resin" include:

(a) aliphatic and cycloaliphatic petroleum hydrocarbon resins

5 having Ring and Ball softening points of from 10 °C to 160 °C, as determined by ASTM method E28-58T, the latter resins resulting from the polymerization of monomers consisting primarily of aliphatic and/or cycloaliphatic olefins and diolefins; also included are the hydrogenated aliphatic and cycloaliphatic petroleum hydrocarbon resins; examples of such commercially available resins based on a C5 olefin fraction of this type are 10 piccotac 95 tackifying resin sold by Hercules Corp. and Escoreze 1310LC sold by ExxonMobil Chemical Company;

(b) Aromatic petroleum hydrocarbon resins and the hydrogenated derivatives thereof;

15 (c) Aliphatic/aromatic petroleum derived hydrocarbon resins and the hydrogenated derivatives thereof;

(d) Aromatic modified cycloaliphatic resins and the hydrogenated derivatives thereof;

20 (e) Polyterpene resins having a softening point of from about 10°C to about 140°C, the latter polyterpene resins generally resulting from the polymerization of terpene hydrocarbons, such as the mono-terpene known as pinene, in the presence of Friedel-Crafts catalysts at moderately low temperatures; also included are the hydrogenated polyterpene resins;

25 (f) Copolymers and terpolymers of natural terpenes, e.g. styrene/terpene,  $\alpha$ -methyl styrene/terpene and vinyl toluene/terpene;

(g) natural and modified rosin such as, for example, gun rosin, wood rosin, tall-oil rosin, distilled rosin, hydrogenated rosin, dimerized rosin and polymerized rosin;

plasticizer may be selected from the group which includes the usual plasticizing oils, such as mineral oil, but also olefin oligomers and low molecular weight polymers, as well as vegetable and animal oils and derivatives of such oils. The petroleum derived oils which may be employed are relatively high boiling materials containing only a minor proportion aromatic hydrocarbons. In this regard, the aromatic hydrocarbons should preferably be less than 30% and more particularly less than 15% of the oil, as measured by the fraction of aromatic carbon atoms. More preferably, the oil may be essentially non-aromatic. The oligomers may be polypropylenes, polybutenes, hydrogenated polyisoprenes, hydrogenated polybutadiens, or the like having average molecular weight between about 350 and about 10,000. Suitable vegetable and animal oils include glycerol esters of the usual fatty acids and polymerization products thereof. Other useful plasticizers can be found in the families of conventional dibenzoate, phosphate, phthalate esters, as well as esters of mono- or polyglycols. Examples of such plasticizers includes, but are not limited to dipropylene glycol dibenzoate, pentaerythritol tetrabenzoate; 2-ethylhexyl diphenyl phosphate, polyethylene glycol 400-di-2-ethylhexoate; butyl benzyl phthalate, dibutyl phthalate and dioctylphthalate. The plasticizers that finds usefulness in the present invention can be any number of different plasticizers but the inventors have discovered that mineral oil and liquid polybutenes having average molecular weight less than 5,000 are particularly advantageous. As will be appreciated, plasticizers have typically been used to lower the viscosity of the overall adhesive composition without substantially decreasing the adhesive strength and/or the service temperature of the adhesive as well as to extend the open time and to improve flexibility of the adhesive.

Waxes can be used to reduce the melt viscosity of the hot melt adhesive composition. Although amounts varying from about 0% to 35% by weight may be used in the composition of the present invention, the preferred amounts are between 0% to 18% by weight. These waxes can also effect the set-up time and the softening point of the adhesive. Among the useful waxes are:

polymers noted above, and thereby the total adhesive system, from the effects of thermal and oxidative degradation which normally occurs during the manufacture and application of the adhesive as well as in the ordinary exposure of the final product to the ambient environment. Among the applicable stabilizers are high molecular weight hindered phenols and multifunction phenols, such as sulfur and phosphorous-containing phenols. Hindered phenols are well known to those skilled in the art and may be characterized as phenolic compounds that also contain sterically bulky radicals in close proximity to the phenolic hydroxyl group thereof. In particular, tertiary butyl groups generally are substituted onto the benzene ring in at least one of the ortho positions relative to the phenolic hydroxyl group. The presence of these sterically bulky substituted radicals in the vicinity of the hydroxyl group serves to retard its stretching frequency and correspondingly, its reactivity; this steric hindrance thus providing the phenolic compound with its stabilizing properties. Representative hindered phenols include:

15        1,3,5-trimethyl-2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl) benzene;  
          pentaerythritol tetrakis-3(3,5-di-tert-butyl-4-hydroxyphenyl) propionate;  
          n-octadecyl-3(3,5-di-tert-butyl-4-hydroxyphenyl) propionate;  
          4,4'-methylenebis(4-methyl-6-tert butylphenol);  
          2,6-di-tert-butylphenol;  
          6-(4-hydroxyphenoxy)-2,4-bis(n-octylthio)-1,3,5-triazine;  
          2,3,6-tris(4-hydroxy-3,5-di-tert-butyl-phenoxy)-1,3,5-triazine;  
          di-n-octadecyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate;  
          2-(n-octylthio)ethyl-3,5-di-tert-butyl-4-hydroxybenzoate; and  
          sorbitol hexa-3(3,5-di-tert-butyl-4-hydroxy-phenyl) propionate.

20        Especially preferred as a stabilizer is pentaerythritol tetrakis-3(3,5-di-tert-butyl-4-hydroxyphenol) propionate.

The performance of these stabilizers may be further enhanced by utilizing, in conjunction therewith; (1) synergists such as, for example, thiodipropionate esters

spiral spray and the like. In a preferred embodiment, the hot melt adhesive is sprayed onto a substrate using spiral spray, which is a preferred technique to produce a filamentary spiral pattern for elastic attachment and construction in diaper manufacturing. In one example, a hot melt coater is equipped with a disc like coating die which has a nozzle tip in the center. The tip is surrounded with a series of inclined orifices for hot air jets to pass through. The hot melt adhesive is pumped out of the nozzle in the form of a fine filament. The filament is then rotated by high-velocity hot air jets coming out of the orifices, thereby producing a helical pattern from a single strand of adhesive. It is not the intent of this invention to provide a full description of spray techniques and the details can be found in the literature.

The adhesive composition of the present invention may be used in a number of applications such as, for example, in disposable nonwoven hygienic articles, paper converting, flexible packaging, wood working, carton and case sealing, labeling and other assembly applications. Particularly preferred applications include disposable diaper and feminine sanitary napkin construction, diaper and adult incontinent brief elastic attachment, diaper and napkin core stabilization, diaper backsheet lamination, industrial filter material conversion, surgical gown and surgical drape assembly, etc.

20

## TESTS AND MATERIALS

Brookfield viscosity was tested according to ASTM D-3236 Method at 350°F.

Ring & Ball softening point was determined with an automated Herzog unit according to ASTM E-28 method.

Peel strength was measured in 180° geometry with a tensile tester (Instron Model 55R1122) in the controlled atmospheric environment (20 °C and 50% relative humidity). Prior to the test, the specimens were conditioned at the controlled environment for approximately 12 hours to ensure the reproducibility

Rexflex RT2280, also available from Huntsman, is an atactic propylene-ethylene copolymer type of APAO having a Brookfiled viscosity of about 8,000 cP at 190 °C, a Tg of about -22 °C and a softening point of about 146 °C.

5 Rexflex RT2780, also available from Huntsman, is an atactic propylene-butene-1 copolymer type of APAO having a Brookfiled viscosity of about 8,000 cP at 190 °C, a Tg of about -23 °C and a softening point of about 110 °C.

10 Eastoflex P1010, obtained from Eastman Chemical Company, Kingsport, TN, is an atactic homopolypropylene type of APAO having a Brookfiled viscosity of about 1,000 cP at 190 °C, a Tg of about -10 °C and a softening point of about 150 °C.

15 Eastoflex M1058, also obtained from Eastman Chemical Company, is an atactic homopolypropylene/copolymer mixture type of APAO having a Brookfiled viscosity of about 5,800 cP at 190 °C and a softening point of about 151 °C.

20 Eastoflex E-1200, also obtained from Eastman Chemical Company, is an atactic propylene-ethylene copolymer type of APAO having a Brookfiled viscosity of about 12,000 cP at 190 °C, a Tg of about -28 °C and a softening point of about 135 °C.

Escorez 5380, available from Exxon, is a very light color, hydrogenated cycloaliphatic hydrocarbon tackifier having an R&B softening point of about 80 °C.

25 Hercotac 1148 is a C5 aliphatic hydrocarbon resin having a R&B softening point of 100 °C. It is available from Hercules Corporation.

Eastotac H130R, available from Eastman Chemical Company, is a partially hydrogenated C5 hydrocarbon resin having a R&B softening point of 130 °C.

30 Sylvaroz ZT 105 is styrenated polyterpene tackifier having a R&B softening point of about 105 °C. It is available from Arizona Chemical Company, Jacksonville, FL.

Kaydal oil is an USP grade white mineral oil plasticizer, obtained from Krompton Corporation, Greenwich, CT.

spiral pattern. The sprayability was deemed acceptable if a good spiral pattern as that depicted in Figure 1b was observed; otherwise, it was deemed unacceptable (Figure 1a). Adhesives were spiral sprayed at 18 g/m<sup>2</sup> coating weight with 0.25 seconds open time and 1 bar compression at the nip rolls and the application temperature was set at 375 °F. The adhesives of Examples of 1 – 3 were found to have almost no room temperature tack, low melt viscosity, good sprayability and good creep retention properties.

TABLE 1. EXAMPLES 1 – 3

Ingredients	Percent (%) by weight		
	1	2	3
Fina EOD 98-05	10.0	-	-
Fina EOD 99-19	-	10.0	22.0
Eastoflex P1010	40.0	-	-
Rexflex RT2280	-	40.0	-
Rexflex RT2180	-	-	15.0
Pennznap 500	10.0	-	25.0
Kaydol Oil	-	10.0	-
Eastotac H130R	39.5	39.5	-
Sylvaros ZT105	-	-	25.0
Marcus 300	-	-	12.5
Antioxidant	0.5	0.5	0.5
Room Temperature Tack	None	None	None
Brookfield Vis. (cP)	3350	9300	7000
R&B Softening Point (°F)	285	288	260
Sprayability	Good	Good	Good
Creep Retention (%)	66	80	72

10

## EXAMPLES 4 - 8

Examples of 4 – 8 were formulated by using the same procedure as herein described above and the ingredients listed in Table 2. These formulations are particularly suited as laminating adhesives for a variety flexible packaging applications and as construction adhesives for disposable nonwoven applications.

15

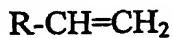
**EXAMPLE 9**

The adhesive in this Example 9 was prepared by using the same procedure herein described above and ingredients consisting of 35.0% by weight Fina EOD 99-19 SPP polymer, 10.0% by weight Eastoflex P1010 APAO, 20.0% by weight Kaydol oil, 34.5% by weight Escorez 5380 tackifier and 0.5% by weight antioxidant. The adhesive sample has a Brookfield Viscosity of 30,700 cP at 375 °F and a softening point of 251 °F. It is particularly useful for case and carton sealing applications. To illustrate the use for such application, reference is made to Figs. 2 and 3 herein where single adhesive beads of about 2 mm in diameter, illustrated as 5 21a and 21b in Figure 2, was applied by hand across the upper surface of top flaps 10 22a and 22b of a corrugated box 20 as shown in the figure. Immediately after the adhesive application, the top flaps 22a and 22b of box 20 were folded over and brought into contact with bottom flaps 23a and 23b to seal the box 20. The flaps 22a, 22b, 23a and 23b were then held together by pressure means for approximately 15 1 minute to seal box 20 as shown in Fig. 3. The adhesive yielded a fiber tearing bond in about 20 minutes after the adhesive application.

Without deviating from the spirit and scope of the present invention, many embodiments and variations can be made by using the components disclosed herein above.

6. The composition of claim 5 wherein the  $\alpha$ -olefin comonomer is selected from the group consisting of ethylene, butene-1 and hexene-1.

7. The composition of claim 1 wherein the APAO polymer is a homopolymer or a copolymer of propylene and at least one  $\alpha$ -olefin comonomer having the following molecular structure:



5 where R is a hydrogen H, or an alkyl or an aryl radical .

8. The composition of claim 7 wherein the  $\alpha$ -olefin comonomer is selected from the group consisting of ethylene, butene-1 and hexene-1.

9. The composition of claim 1 wherein the tackifier is selected from the group consisting of aliphatic and cycloaliphatic hydrocarbon resins and their hydrogenated derivatives, aromatic and hydrogenated aromatic hydrocarbon resins, aromatically modified aliphatic or cycloaliphatic resins and their hydrogenated derivatives, polyterpene and styrenated polyterpene resins.

5 10. The composition of claim 9 wherein the tackifier has a R&B softening point equal to or greater than 80 °C.

11. The composition of claim 10 wherein the tackifier is a C-5 aliphatic hydrocarbon resin.

12. The composition of claim 1 wherein the plasticizer is selected from the group consisting of mineral oil and liquid polybutene.

13. The composition of claim 12 wherein the mineral oil has less than 30% aromatic carbon atoms.

14. The composition of claim 1 wherein the adhesive composition further comprises a wax in the amount up to 30% by weight.

15. The composition of claim 14 wherein said wax is selected from the group consisting of petroleum waxes, low molecular weight polyethylene and polypropylene, synthetic waxes and polyolefin waxes.

1 / 2

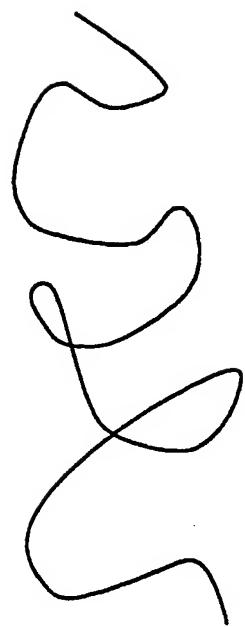


FIG. 1(a)

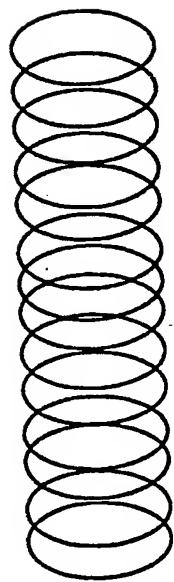


FIG. 1(b)

## INTERNATIONAL SEARCH REPORT

International Application No  
PCT/US 02/26297

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 7 C09J123/10

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 C09J C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4 998 928 A (MALETSKY ALBERT ET AL) 12 March 1991 (1991-03-12) column 1, line 58 -column 2, line 5 ---	1-21
A	EP 0 931 814 A (FINA RESEARCH) 28 July 1999 (1999-07-28) example 21; table 9 ---	1-21
A	PATENT ABSTRACTS OF JAPAN vol. 2000, no. 10, 17 November 2000 (2000-11-17) & JP 2000 191852 A (MITSUI CHEMICALS INC), 11 July 2000 (2000-07-11) abstract ---	1-21
A	WO 94 04625 A (EXXON CHEMICAL PATENTS INC) 3 March 1994 (1994-03-03) examples 6-13 ---	1-21
	-/-	

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

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PCT/US 02/26297

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